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Study on defects in H^+ ion implanted B2 type Fe-Al alloy using slow positron beamS. Komagata¹, A. Kawasuso², A. Yabuuchi², M. Maekawa², C. Batchulun³, K. Yasuda³,
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Abstract

Fe48-at.%Al alloy were implanted with 50 keV H^+ ions to the fluence of 3×10^{16} and 1×10^{18} /cm² at room temperature. Positron annihilation Doppler broadening and lifetime measurements for these alloys have been carried out using slow positron beam apparatus with an energy range of 0.2 to 30.2 keV. The positron annihilation S-parameter decreased by H^+ ion irradiation. Also the positron lifetimes for hydrogen deposited region in the alloy decreased by the irradiation. These results show that implanted H atoms were trapped by vacancy type defects.

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1. Introduction

B2 type Fe-Al alloys are potential structural materials for high temperature applications. The importance of the Fe-Al alloys is due to their excellent oxidation and sulphidation resistance. A low density of the Fe-Al alloys and the price of their components are the additional advantages of these materials. The main obstacle in the practical usage of Fe-Al alloys is their low plasticity at room temperature due to internal weakness of grain boundaries, excess of thermal vacancies and so on [1]. Sensitivity to H that causes so-called environmental embrittlement is one of the main reasons for low plasticity and for susceptibility to brittle fracture of the alloys at room temperature [2, 3]. It is known that B2 type Fe-Al alloys are capable to include a large number of thermal vacancies, the concentration of which is the order of atomic % at room temperature [4]. The effects of H atoms on vacancy properties in Fe-Al

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alloys have received a considerable attention in scientific and technologic fields. However, quantitative characterization has never been provided on the states of vacancy-type defects with hydrogen atoms in these alloys.

To study the interaction between vacancy-type defects and H atoms, we have to introduce H atoms into samples. The electrolytic H charging method is most popular method to study the interaction between vacancy type defects and H atoms [5]. However this technique cannot well control the amount of hydrogen atoms in the samples. In contrast, The H^+ ion implantation technique can control the total number and the implantation depth of H^+ ions by changing the ion energy and the total fluence. Positron annihilation technique is an excellent way, especially for detecting vacancy-type defects in materials. Some experiments using this technique have already been reported for B2 Fe-Al alloys [6–9]. Diego et al. reported positron lifetimes for several types of defects in B2 type Fe-Al, such as the values of the lifetime are 181 ps for Al-vacancy (V_{Al}) and 189 ps for Fe-vacancy (V_{Fe}) [9]. However, the interaction between vacancy-type defects and H atoms in Fe-Al alloys using positron annihilation measurement has never been fully studied so far. In this study, the slow positron beam technique was used to measure the interaction of H atoms with vacancy-type defects in Fe-Al alloy. We prepared Fe-48at.%Al alloy with a single B2 structure at room temperature.

2. Experimental Procedure

Fe-48 at.%Al alloys were prepared from aluminum with 99.999% purity and iron with 99.99% purity by arc melting method under an argon gas atmosphere. They were cut into thin sheets with the dimension of 10mm x 10mm x 1.0 mm. The surface of each alloy was mechanically polished. Each sheet was heat-treated at 1273 K for 2 h followed by slow-cooling to room temperature in vacuum. The alloys were confirmed to have a single B2 phase by an X-ray diffraction (XRD) measurement.

The samples were irradiated with 50 keV H^+ ions to the fluence of $3 \times 10^{16} / \text{cm}^2$, $3 \times 10^{17} / \text{cm}^2$ and $1 \times 10^{18} / \text{cm}^2$, at room temperature at Wakasa Wan Energy Research Center (WERC). The damage profile and the profile of implanted H atoms were calculated using SRIM 2006 code [10]. The calculation shows that the implanted hydrogen atoms are distributed around the depth of 320nm from the sample surface. The SRIM calculation also shows that the average numbers of atomic displacements (displacements per atom) at the depth from 200 nm to 400 nm are 7.8×10^{-2} dpa for the fluence of $3 \times 10^{16} / \text{cm}^2$, 7.8×10^{-1} dpa for the fluence of $3 \times 10^{17} / \text{cm}^2$ and 2.6 dpa for the fluence of $1 \times 10^{18} / \text{cm}^2$. The average H concentration at the depth from 200 ~ 400 nm are 1.7 at.% for the fluence of $3 \times 10^{16} / \text{cm}^2$ and 57 at.% for the fluence of $1 \times 10^{18} / \text{cm}^2$.

Positron annihilation lifetime and Doppler broadening measurements were performed for the unirradiated Fe-Al samples and the H^+ ion-irradiated samples at JAEA-Takasaki, Japan. The Doppler broadening of the annihilation line was measured for the positron energy range from 0.2 keV to 30.2 keV corresponding to the positron incident depth from 0.5 to 1.6×10^3 nm from the surface [11]. The Doppler broadening S parameter was determined in the usual way from the spectra of annihilation gamma ray [12]. The window for each analyzed value of S-parameter was defined as $511 \text{ keV} \pm 970 \text{ eV}$. The positron lifetime for each alloy was also measured by the positron beam with the fixed energy of 10 keV. The positron lifetime measurement was carried out with a time resolution of about 250 ps. The statistical error of the fits was 2 ps. Total measuring counts for each measurement were 5×10^5 for Doppler broadening and 5×10^5 for positron lifetime.

3. Results and Discussion

The positron annihilation curve for the unirradiated alloy consists of only one component with the lifetime of 199 ps, which is larger than the value for the annihilation in matrix. This result suggests that the most positrons are annihilated at single or di-vacancies. Moreover, the vacancy concentration can be estimated as more than the order of 100 at.ppm which is estimated by using two state trapping model with specific trapping rate of $10^{15}/\text{s}$ for vacancy in bcc Fe [13]. This value is consistent with the result of the previous report which shows that about 1at.% vacancies exist in same alloy [8]. Figure 1 shows the positron annihilation S-parameter of Fe-48at.%Al alloy before implantation as a function of incident positron energy. Their values are nearly constant irrespective of positron

energy. A large value of the S parameter also shows that vacancy-type defects have already existed before implantation. This result shows that vacancy-type defects with the similar size are distributed through the sample.

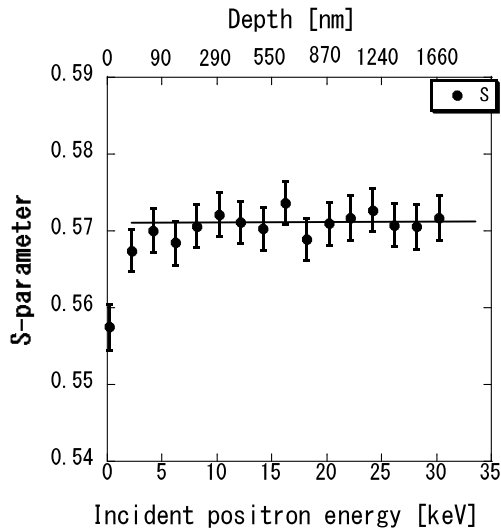


Fig. 1. S-parameter as a function of incident positron energy, for Fe-48at.%Al alloy before irradiation.

Figs. 2 (a) and (b) show the S-parameter for H irradiated samples as a function of positron energy and corresponding depth. In the figures, the distribution of implanted H atoms, which has been calculated by SRIM code, is also shown.

In the H^+ ion implanted alloys, the S-parameter is not constant against the positron energy or corresponding depth, but a dip can be observed around the depth of 300 nm, where the implanted H atoms accumulate. H^+ ion induced defects are also expected to localize around this region. If the accumulating defects contributed to the change in S-parameter, they would increase the S-parameter. The experimental result, however, shows the opposite case. To

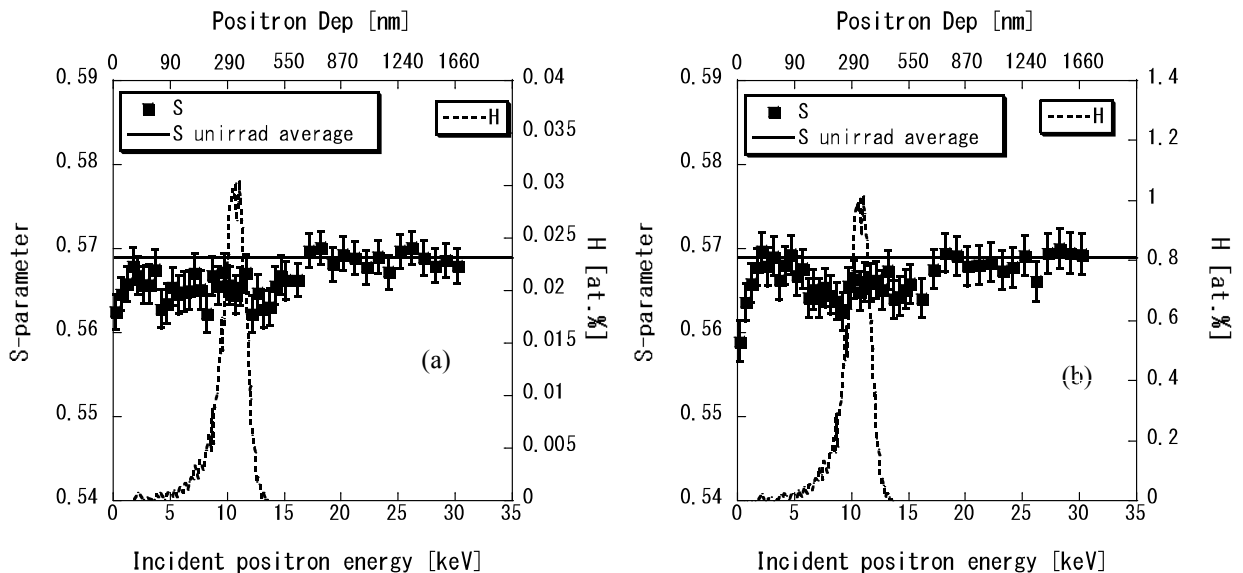


Fig. 2. S-parameter for the Fe-48at.%Al samples implanted with H^+ ions to the fluence of (a) $3 \times 10^{16} / \text{cm}^2$ and (b) $1 \times 10^{18} / \text{cm}^2$, as a function of incident positron energy and corresponding depth. Also shown is the H^+ implantation profile.

explain the result, we have to consider the effect of implanted H atoms on S-parameter.

Fig. 3 also clearly shows the effect of implanted H atoms on the S-parameter. The figure plots the value of S-parameter as a function of H^+ fluence. In the region where H atoms do not accumulate, the S-parameter is about the same as that before implantation. In contrast, the decrease in S-parameter is observed in the region where hydrogen atoms mainly accumulate. The present experimental result can be explained tentatively as follows; in the unirradiated samples, the positron annihilation occurs in the already-existing vacancy-type defects. In the H^+ implanted samples, implanted hydrogen atoms are trapped by vacancy-type defects, causing the increase in electron density in vacancy-type defects, and the resulting decrease in S-parameter can be observed. On the other hands, the S-parameter does not change with H^+ implantation, although the concentration of vacancy and hydrogen atom increases with increasing irradiation fluence. In this case, it is considered that most implanted hydrogen atoms might be trapped by vacancies, and surplus hydrogen atoms would reach to the surface or localized at interstitial site of which is not attractive for positron. We can not discuss where the non-trapped H atoms are, precisely. Positron trapping at this vacancy-hydrogen pair is already saturated even at $3 \times 10^{16} / \text{cm}^2$, so that the S-parameter remains constant seemingly. The effect of implanted hydrogen atoms is also observed as the decrease in positron lifetime. Fig. 4 shows that the lifetime of positron decreases by 8 ps after the hydrogen implantation. This result is consistent with the result of Doppler broadening measurement.

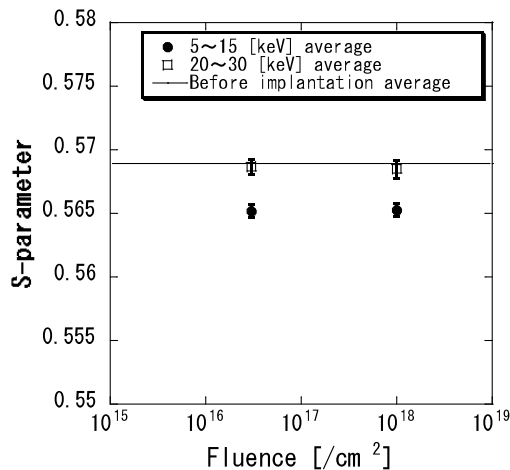


Fig. 3. Average of S-parameter for 50 keV H^+ implanted Fe-48at.%Al samples. Open squares S-parameter values measured by 20-30keV positron beam, solid circles: S-parameter values measured by 5-15 keV positron beam.

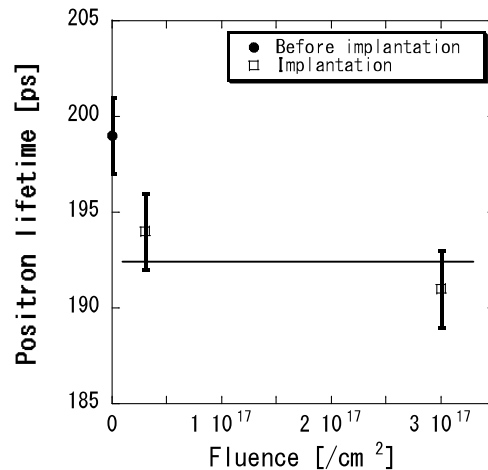


Fig. 4. Positron lifetime for the before implantation Fe-48at.%Al alloy and 50keV H^+ implanted Fe-48at.%Al alloy.

4. Conclusions

Defects in H^+ ion implanted Fe-Al alloys were studied by using a slow positron beam. The Doppler broadening S-parameter decreased by H^+ ion implantation, Also the positron lifetime for H implanted region in this alloy was about 8 psec lower than that before implantation. These results suggest the interaction between H atoms and vacancy type defects. The implanted H atoms were trapped by vacancy type defects in Fe-Al alloy.

Acknowledgements

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References

- [1] Pike, L.M., Liu, C.T., *Scr. Mater.* 1998 38:1475.
- [2] Stoloff, N.S., Liu, C.T., *Intermetallics* 2 1994 75.
- [3] Vyas, S., Viswanathan, S., Sikka, V.K., *Scr. Metall. et Mater.* 1992 27:185.
- [4] Kogachi, M., Haraguchi, T., *Mater. Sci. and Eng.* 1997 A230:124-131.
- [5] Barcik, J., St"pie#, K., *Mater. Sci. and Eng.* 2002 A334:28–32.
- [6] Würschum, R., Grupp, C., Schaefer, H-E, *Phys. Rev. Lett.* 1995 75:97.
- [7] Wolff, J., Franz, M., Broska, A., Kerl, R., Weinhausen, M., Köhler, B., Brauer, M., Faupel, F., Hehenkamp, Th., *Intermetallics* 7 1999 289–300.
- [8] Haraguchi, T., Hori, F., Oshima, R., Kogachi, M., *Intermetallics* 9 2001 763–770.
- [9] de Diego, N., Plazaola, F., Jime'nez, J.A., Serna, J., del Río, J., *Acta Mater.* 2005 53:163–172.
- [10] <http://www.srim.org/>
- [11] Valkealahti, S., and Nieminen, R. M., *Appl. Phys.* 1983 A32:95.
- [12] MacKenzie, I.K., Eady, J.A., Gingerich, R.R., *Phys. Lett.* 1970 33A:5279.
- [13] Hautajarvi, P., Heino, J., Manninen, M., *Phil. Mag.* 1977 vol.35, 4:973-981.